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HOT MELT ADHESIVE COMPOSITION [Hotto Meruto Setchakuzai Soseibutsu]

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/2\* [Claims]

[Claim 1] A hot melt adhesive composition that is comprised of an ethylene- $\alpha$ ·olefin copolymer having a density of 0.90 g/cm<sup>3</sup> or lower, a styrene-ethylene-propylene-styrene block copolymer, and a tackifying resin.

[Claim 2] The hot melt adhesive composition stated in Claim 1, in which the content of the aforesaid ethylene-α·olefin copolymer is from 10 to 80 % by weight, the content of the aforesaid styrene-ethylenepropylene-styrene block copolymer is from 1 to 50 % by weight, and the content of the aforesaid tackifying resin is from 19 to 60 % by weight.

[Claim 3] The hot melt adhesive composition stated in Claim 1 or 2, in which the content of styrene in the aforesaid styrene-ethylenepropylene-styrene block copolymer is from 10 to 35 % by weight.

[Claim 4] The hot melt adhesive composition stated in any of Claims 1 through 3 that further contains a plasticizing oil. [Detailed Description of the Invention]

[0001] [Technical Field of the Invention]

The present invention pertains to a hot melt adhesive composition that is ideal for use in, for example, the assembly of sanitary articles, such as disposable diapers, catamenial pads, etc. [0002] [Prior Art and Problems that the Invention Intends to Solve]

To impart a better effect of preventing stuffiness and skin irritation to sanitary articles, such as disposable diapers,

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catamenial pads, in particular, disposable diapers (a) that employ porous polyethylene film as the back sheet (outer sheet) and (b) those that use inner leg gathers, which are called three-dimensional gathers, have been proposed and are commonly used. The disposable diapers of the aforesaid "a" type are typically constructed by using a nonwoven cloth as the top sheet (the inner sheet that makes contact with the skin) and by bonding said top sheet to the back sheet by means of a hot melt adhesive. The disposable diapers of the aforesaid "b" type are constructed by bonding a three-dimensional gather (which is prepared by wrapping an elastic material with a nonwoven cloth) and the aforesaid top sheet and/or back sheet by means of a hot melt adhesive. The aforesaid hot melt adhesive used for the aforesaid disposable diapers "a" and "b" is, due to the nature of disposable diapers, required to satisfy the following conditions as important qualities: (1) the adhesive should not exude the low molecular substances contained in it to the porous polyethylene film used as the back sheet and mar the appearance; (2) in the case of using it for the assembly of the aforesaid three-dimensional gather, the adhesive should not cause skin irritation even if it makes direct contact with the skin; (3) the adhesive should be soft, white or colorless, and odorless; and so forth.

[0003] As the aforesaid hot melt adhesive, it is a common practice to use synthetic rubber hot melt adhesives that use synthetic rubber as the base polymer. These synthetic rubber hot melt adhesives

have tackiness even at room temperature and are in a sticky state, so to speak, but, because the quantity of a low molecular substance that can be incorporated into said hot melt adhesives is extremely larger, compared with those of adhesives that use other polymers, they are capable of preventing low molecular substances from seeping through porous polyethylene film used for stuffiness prevention. In recent years, however, for the purpose of making disposable diapers feeling softer and also for the purpose of cutting cost, the aerial densities (weight) of the aforesaid porous ethylene film and nonwoven cloth, which are the constituent materials of disposable diapers, have been reduced, and, under such a condition with reduced aerial densities of the base materials, the aforesaid synthetic rubber hot melt adhesives are not capable of inhibiting the seepage. As a result, disposable diapers that are produced using conventional synthetic rubber hot melt adhesives have the problem of poor appearance caused by the seeping out of the adhesives or the problem of seeping out onto the skin side and making contact with the skin of the user, thus causing skin irritation. In other words, they have the problem of not being able to satisfy the aforesaid conditions 1 and 2.

[0004] As the method for preventing the aforesaid seeping problem, reducing the blending amount of the low molecular substances in the hot melt adhesives has been considered. This, however, causes the adhesive force to decrease, thus presenting the problem of not being able to obtain the desired adhesion strength. Consideration has also

been given to, for example, increasing the viscosity of the adhesives so as to prevent the adhesives from coming out through the space between fibers, but this idea has not reached a satisfactory level yet. Commonly used as less sticky adhesives are EVA-based or  $\alpha$ -olefin-based hot melt adhesives, which have an ethylene-vinyl acetate copolymer (EVA) or ethylene- $\alpha$ -olefin copolymer as the base polymer, to which is added a tackifying resin or wax. This type of hot melt adhesive exhibits tackiness when melted, and, because the base polymer has crystallinity, the base polymer, when cooled after the adhesion, crystallizes (that is, it solidifies into a stiff condition) and loses tackiness. Therefore, after it is cooled, the adhesive has the characteristic of not causing skin irritation even if it makes direct contact with the skin. However, because it feels hard after it has crystallized, it has a problem in that disposable diapers proper become hard and could rub against the skin of the user and cause skin irritation.

[0005] To achieve a softer texture, the addition of a plasticizer in liquid form has been proposed, but, if the aforesaid liquid form plasticizer is added to a hot melt adhesive having the aforesaid base polymer, tackiness remains even after the adhesive is cooled, and the adhesive adversely affects the skin, etc., during use.

[0006] In short, the hot melt adhesives that have been proposed heretofore cannot satisfy all of the aforesaid requirements (1) through (3), and there has been a demand for the development of a hot melt adhesive that can meet all of the aforesaid requirements (1) through (3).

[0007] Accordingly, the objective of the present invention is to provide a hot melt adhesive composition that does not cause the seeping of low molecular substances, that loses tackiness after it is cooled and solidified, and that, when used as the adhesive for assembling sanitary articles, such as disposable diapers, etc., does not impair the texture of said disposable diapers.

[0008] [Means Of Solving The Problems]

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The present invention achieved the aforesaid objective by providing a hot melt adhesive composition that contains an ethylene- $\alpha$ -olefin copolymer whose permanent set is 50 % or less and whose hysteresis ratio is 5.0 or less, a styrene-ethylene-propylene-styrene block copolymer, and a tackifying resin.

[0009] [Mode of Implementing the Invention]

The following explains the hot melt adhesive composition of the present invention in further detail. The hot melt adhesive composition of the present invention is characterized by containing a specific ethylene-lpha·olefin copolymer, styrene-ethylene-propylene-styrene block copolymer (hereinafter referred to as a "SEPS"), and tackifying resin.

[0010] The aforesaid specific ethylene- $\alpha$ -olefin copolymer used in the hot melt adhesive composition of the present invention is an ethylene- $\alpha$ -olefin copolymer whose density is 0.90 g/cm $^3$  or lower, preferably from 0.880 to 0.860 g/cm3. If the aforesaid density exceeds

0.90 g/cm<sup>3</sup>, the melt viscosity becomes too high for a hot melt adhesive. Here, the aforesaid density is determined in a manner similar to a method ordinarily used for measuring the density of known polymers.

[0011] The aforesaid ethylene- $\alpha$ -olefin copolymer is not specifically limited as long as it satisfies the aforesaid density, but the present invention preferably uses an ethylene- $\alpha$ -olefin copolymer that is obtained by copolymerizing the components with the use of a cyclopentadienyl complex as the catalyst.

[0012] With respect to the aforesaid ethylene- $\alpha$ -olefin copolymer, the  $\alpha$ -olefin that is copolymerized with ethylene is an  $\alpha$ -olefin having from 3 to 30 carbon atoms, concrete examples of which include propylene, 1-butene, 1-pentene, 1-hexene, 1-octene, 1-heptene, 4-methyl pentene-1, 4-methyl hexene-1, 4,4,-dimethyl pentene-1, octadecene, etc. Of these, 1-hexene, 1-octene, 1-heptene, and 4-methyl-pentene-1 are preferably used.

[0013] Examples of the aforesaid cyclopentadienyl complex used as the catalyst in the process of copolymerizing ethylene and  $\alpha$ -olefin to form the aforesaid ethylene- $\alpha$ -olefin copolymer include compounds represented by Formula I below:

[0014]  $ML_X$  (I)

[In the formula, M indicates a transition metal selected from a group consisting of Zr, Ti, Hf, V, Nb, Ta, and Cr; L is a ligand that coordinates to said transition metal and represents a group having a

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group having a cyclopentadienyl group.]

cyclopentadienyl backbone, a hydrocarbon group having 1 to 12 carbon atoms, an alkoxy group having 1 to 12 carbon atoms, an aryloxy group having 1 to 12 carbon atoms, a trialkyl silyl group having 1 to 12 carbon atoms, an  $SO_3R$  group (in which R stands for a  $C_{1-8}$  hydrocarbon group that may optionally have a substituent, such as a halogen atom, etc.), a halogen atom, or a hydrogen atom; and x is the same number as the atomic valence of the aforesaid transition metal. When more than one L coordinate to the metal, they may be different from one another, but a minimum of one should be a group having a cyclopentadienyl backbone. That is, when the aforesaid x is 1, the aforesaid L is a group having a cyclopentadienyl backbone, and, if the aforesaid x is 2 or larger, a minimum of one among a plurality of the aforesaid Ls is a

[0015] Some examples of the aforesaid group having a cyclopentadienyl backbone include a cyclopentadienyl group; alkylsubstituted cyclopentadienyl groups, such as a methyl cyclopentadienyl group, dimethyl cyclopentadienyl group, trimethyl cyclopentadienyl group, tetramethyl cyclopentadienyl group, pentamethyl cyclopentadienyl group, ethyl cyclopentadienyl group, methyl ethyl cyclopentadienyl group, propyl cyclopentadienyl group, methyl propyl cyclopentadienyl group, butyl cyclopentadienyl group, methyl butyl cyclopentadienyl group, hexyl cyclopentadienyl group, etc.; an indenyl group; a 4,5,6,7-tetrahydroindenyl group; a fluorenyl group; etc.

These groups may optionally be substituted with halogen atoms, trialkyl silyl groups, etc.

[0016] As the aforesaid group having a cyclopentadienyl backbone, the present invention preferably uses alkyl-substituted cyclopentadienyl groups among the examples listed in the foregoing.

[0017] If the compound represented by the aforesaid General Formula 1 contains two or more groups having a cyclopentadienyl backbone, two groups having a cyclopentadienyl backbone among them may optionally be bonded with each other by means of an alkylene group, such as an ethylene group, propylene group, etc.; a substituted alkylene group, such as isopropylidene, diphenyl methylene, etc.; a silylene group; or a substituted silylene group, such as a dimethyl silylene group, diphenyl silylene group, methyl phenyl silylene group, etc.

[0018] Examples of the aforesaid hydrocarbon group having 1 to 12 carbon atoms include an alkyl group, cycloalkyl group, aryl group, aralkyl group, etc. More specifically, examples of the alkyl group include a methyl group, ethyl group, propyl group, isopropyl group, butyl group, etc. Examples of the cycloalkyl group include a cyclopentyl group, cyclohexyl group, etc., and examples of the aryl group include a phenyl group, tolyl group, etc. Examples of the aralkyl group include a benzyl group, neophyl group, etc. Examples of the alkoxy group include a methoxy group, ethoxy group, butoxy group, etc., and examples of the aryloxy group include a phenoxy group, etc.

Examples of the halogen atom include a fluorine atom, chlorine atom, bromine atom, iodine atom, etc.

[0019] Examples of the aforesaid SO<sub>3</sub>R group include a p-toluene /4 sulfonate group, methane sulfonate group, trifluoromethane sulfonate group, etc.

[0020] When the atomic valence of the transition metal is, for example, 4, the compound that contains this type of group having a cyclopentadienyl backbone is represented more specifically by the following General Formula II below.

[0021]  $R^{2}_{k}$   $R^{3}_{1}$   $R^{4}_{m}$   $R^{5}_{n}$  M (II)

[In the formula, M represents the aforesaid transition metal; R<sup>2</sup> is a group (ligand) having a cyclopentadienyl backbone; R3, R4, and R5 each independently represent a group having a cyclopentadienyl backbone, alkyl group, cycloalkyl group, aryl group, aralkyl group, alkoxy group, aryloxy group, trialkyl silyl group, SO3R group, halogen atom, or hydrogen atom; k is an integer of 1 or larger; and k + l + m + n = 4

[0022] Among the compounds represented by the aforesaid Formula II, the present invention preferably uses a compound in which a minimum of two among  $R^2$ ,  $R^3$ ,  $R^4$ , and  $R^5$ --for example,  $R^2$  and  $R^3$ --in the aforesaid formula are groups (ligands) having a cyclopentadienyl backbone, and these groups (for example, R2 and R3) having a cyclopentadienyl backbone may be bonded as described in the foregoing.

[0023] The following lists concrete examples of the aforesaid compound in which M stands for zirconium: bis (indenyl) zirconium

dichloride, bis (indenyl) zirconium dibromide, bis (indenyl) zirconium bis (p toluene sulfonate) [sic] bis 4,5,6,7-tetrahydroindenyl) zirconium dichloride, bis (fluorenyl) zirconium dichloride, ethylene bis (indenyl) zirconium dichloride, ethylene bis (indenyl) zirconium dibromide, ethylene bis (indenyl) dimethyl zirconium, ethylene bis (indenyl) diphenyl zirconium, ethylene bis (indenyl) methyl zirconium monochloride, ethylene bis (indenyl) zirconium bis (methane sulfonate), ethylene bis (indenyl) zirconium bis (p-toluene sulfonate), ethylene bis (indenyl) zirconium bis (trifluoromethane sulfonate), ethylene bis (4,5,6,7-tetrahydroindenyl) zirconium dichloride, isopropylidene (cyclopentadienyl-fluorenyl) zirconium dichloride, isopropylidene (cyclopentadienyl-methyl cyclopentadienyl) zirconium dichloride, dimethyl silylene bis (cyclopentadienyl) zirconium dichloride, dimethyl silylene bis (methyl cyclopentadienyl) zirconium dichloride, dimethyl silylene bis (dimethyl cyclopentadienyl) zirconium dichloride, dimethyl silylene bis (trimethyl cyclopentadienyl) zirconium dichloride, dimethyl silylene bis (indenyl) zirconium dichloride, dimethyl silylene bis (indenyl) zirconium bis (trifluoromethane sulfonate), dimethyl silylene bis (4,5,6,7-tetrahydroindenyl) zirconium dichloride, dimethyl silylene bis (cyclopentadienylfluorenyl) zirconium dichloride, diphenyl silylene bis (indenyl) zirconium dichloride, methyl phenyl silylene bis (indenyl) zirconium dichloride, bis (cyclopentadienyl) zirconium dichloride, bis (cyclopentadienyl) zirconium dibromide, bis (cyclopentadienyl) methyl

zirconium monochloride, bis (cyclopentadienyl) ethyl zirconium monochloride, bis (cyclopentadienyl) cyclohexyl zirconium monochloride, bis (cyclopentadienyl) phenyl zirconium monochloride, bis (cyclopentadienyl) benzyl zirconium monochloride, bis (cyclopentadienyl) zirconium monochloride monohalide, bis (cyclopentadienyl) methyl zirconium monohalide, bis (cyclopentadienyl) dimethyl zirconium, bis (cyclopentadienyl) diphenyl zirconium, bis (cyclopentadienyl) dibenzyl zirconium, bis (cyclopentadienyl) zirconium methoxy chloride, bis (cyclopentadienyl) zirconium ethoxy chloride, bis (cyclopentadienyl) zirconium bis (methane sulfonate), bis (cyclopentadienyl) zirconium bis (p-toluene sulfonate), bis (cyclopentadienyl) zirconium bis (trifluoromethane sulfonate), bis (methyl cyclopentadienyl) zirconium dichloride, bis (dimethyl cyclopentadienyl) zirconium dichloride, bis (dimethyl cyclopentadienyl) zirconium ethoxy chloride, bis (dimethyl cyclopentadienyl) zirconium bis (trifluoromethane sulfonate), bis (ethyl cyclopentadienyl) zirconium dichloride, bis (methyl ethyl cyclopentadienyl) zirconium dichloride, bis (propyl cyclopentadienyl) zirconium dichloride, bis (methyl propyl cyclopentadienyl) zirconium dichloride, bis (butyl cyclopentadienyl) zirconium dichloride, bis (methyl butyl cyclopentadienyl) zirconium dichloride, bis (methyl butyl cyclopentadienyl) zirconium bis (methane sulfonate), bis (trimethyl cyclopentadienyl) zirconium dichloride, bis (tetramethyl cyclopentadienyl) zirconium dichloride, bis (pentamethyl

cyclopentadienyl) zirconium dichloride, bis (hexyl cyclopentadienyl) zirconium dichloride, and bis (trimethyl silyl cyclopentadienyl) zirconium dichloride.

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[0024] In the aforesaid compounds listed as examples, the disubstitution of a cyclopentadienyl ring, such as dimethyl cyclopentadienyl, etc., include 1,2- and 1,3-substitutions, and the trisubstitution of dimethyl cyclopentadienyl, etc., include 1,2,3-and 1,2,4-substitutions. Furthermore, alkyl groups, such as a propyl group, butyl group, etc., include n-, i-, sec-, tert-, etc., isomers. Examples of the aforesaid compound further include cyclopentadienyl complexes resulting from replacing zirconium in the compounds cited in the foregoing with titanium, hafnium, vanadium, niobium, tantalum, or chromium.

[0025] The aforesaid cyclopentadienyl complexes can be used singly or as a mixture. They can be diluted in a hydrocarbon or halogenated hydrocarbon for use. In particular, the present invention preferably uses a zirconosene compound that has zirconium as the center metal atom and that has a minimum of two groups having a cyclopentadienyl backbone as the ligand.

[0026] The aforesaid cyclopentadienyl complexes can be used concomitantly with ordinary aluminoxane compounds or with compounds that react with said cyclopentadienyl complexes and form stable anions.

[0027] The present invention can also employ, as the cyclopentadienyl complex catalyst, polymerization catalysts that are described in JP-A-H04-253711, JP-A-H04-279592, PCT-H06-503585, JP-A-H03-188092, and JP-A-H02-84407.

[0028] The compositional ratio of ethylene to  $\alpha$ -olefin in the aforesaid ethylene- $\alpha$ -olefin copolymer is preferably from 40 to 98 % by weight of ethylene to from 60 to 2 % by weight of  $\alpha$ -olefin.

[0029] The aforesaid ethylene- $\alpha$ ·olefin copolymer is prepared by polymerizing ethylene and  $\alpha$ ·olefin in the presence of a cyclopentadienyl complex by high-pressure ionic polymerization under from 100 to 3,000 kg/cm², preferably from 300 to 2,000 kg/cm², at a temperature of from 125 to 250 °C, preferably from 150 to 200 °C. Especially when the aforesaid cyclopentadienyl complex and the aforesaid aluminoxane are used concomitantly for the polymerization, the polymerization is carried out according to the method described in JP-A-S61-130314, JP-A-S60-35006, JP-A-S58-19309, JP-A-S60-35008, JP-A-H03-163088, or the like. When the polymerization is carried out using the aforesaid cyclopentadienyl complex concomitantly with a compound that reacts with said cyclopentadienyl complex and forms a stable anion, the polymerization is carried out according to the high-pressure ionic polymerization method described in EPO 277,004, International Publication WO 92/01723, or the like.

[0030] The MFR of the aforesaid ethylene- $\alpha$ -olefin copolymer is preferably 0.5 to 500 g/10 minutes (as measured pursuant to JIS K-7210 at 190 °C with a 2.16 kg load.)

[0031] The permanent set of the aforesaid ethylene- $\alpha$ -olefin copolymer is preferably 50 % or less, better yet, from 0 to 30 %, and its hysteresis ratio is preferably 5.0 or less, better yet, from 1.0 to 3.5. If the aforesaid permanent set exceeds 50 % or the aforesaid hysteresis ratio exceeds 5.0, the resulting hot melt adhesive tends to have high melt viscosity, which is not desirable.

[0032] The aforesaid permanent set and the aforesaid hysteresis ratio are determined as follows.

Method for determining permanent set: The length of a sample that, in the same manner as in the following method for measuring a hysteresis ratio, has been stretched 100 % and subsequently released back to the initial chuck-to-chuck distance is measured, and the proportion (%) of the length that cannot be relaxed (the length of the portion that has been stretched and stayed longer than the initial length) after the 100 % stretching to the initial length (the initial chuck-to-chuck distance) is found and taken as the permanent set.

Method for determining hysteresis ratio: In order to evaluate the flexibility of each sample, its hysteresis ratio is measured using an elongation tester equipped with an accumulator. More specifically, a 25 mm-wide sample is prepared from the aforesaid elastic component, and said sample is fastened to the aforesaid tester so as to set the chuck-to-chuck distance to 100 mm. Thereafter, each sample is stretched 100 % at a speed of 300 mm/minute, thereby preparing a stretching curve, after which the sample is relaxed at the same speed

until the chuck-to-chuck distance returns to the original length, 100 mm, thereby preparing a relaxation curve. The area under the stretching curve and the area under the relaxation curve are measured, and these are assigned to the following equation to calculate the hysteresis ratio.

Hysteresis Ratio = Area under Stretching Curve / Area Under Relaxation Curve

[0033] The aforesaid SEPS is a component that can contain low molecular substances in an extremely large quantity and, consequently, is capable of inhibiting the seepage of said low molecular substances, and it has a structure in which an ethylene unit and propylene unit are present between the styrene polymer blocks located at both ends. Furthermore, because the aforesaid SEPS has the same backbone as the ethylene- $\alpha$  olefin copolymer from the standpoint of chemical structure, it is more miscible with the aforesaid ethylene- $\alpha$ -olefin copolymer than other synthetic rubber, such as a styrene-isoprene-styrene block copolymer (SIS), styrene-butadiene-styrene block copolymer (SBS), styrene-ethylene-butylene-styrene copolymer (SEBS), etc., and is capable of dramatically improving the low-temperature adhesion property of the resulting adhesive composition.

[0034] It is preferable to set the content of the styrene in the aforesaid SEPS to the range of from 10 to 35 % by weight. If the aforesaid content is less than 10 % by weight, tackiness remains even after the composition is cooled, whereas a content exceeding 35 % by /6

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weight is also not desirable because the low-temperature adhesion property tends to deteriorate.

[0035] The aforesaid SEPS can be obtained, for example, by hydrogenating the isoprene polymer moiety of a styrene-isoprenestyrene block copolymer, which has an isoprene polymer block between styrene polymer blocks at both ends.

[0036] As the aforesaid tackifying resin used in the present invention, those that are used for ordinary hot melt adhesive compositions can be used with no specific limitation. However, the present invention preferably uses hydrogenated resins, such as hydrogenated aromatic petroleum resins, hydrogenated aliphatic petroleum resins, hydrogenated terpene resins, etc., because the hydrogenation dramatically reduces double bonds from the standpoint of chemical structure, and, as a result, these resins have excellent heat stability and do not pose the problem of hue changes or bad odor caused by deterioration over time. Furthermore, because this type of tackifying resin is highly miscible with the aforesaid ethylene- $\alpha$ -olefin copolymer and SEPS, its use does not affect the adhesion property adversely. The softening temperature of the aforesaid tackifying resin is not specifically limited, and it may be either solid or liquid at room temperature, but the liquid type is preferable because it can function as a plasticizer.

[0037] It is preferable for the hot melt adhesive composition of the present invention to contain the aforesaid ethylene- $\alpha$ -olefin

copolymer in a quantity of from 10 to 80 % by weight, better yet, from 15 to 50 % by weight; the aforesaid styrene-styrene [sic] -propylenestyrene block copolymer in a quantity of from 1 to 50 % by weight, better yet, from 5 to 30 % by weight; and the aforesaid tackifying resin in a quantity of from 19 to 60 % by weight, better yet, from 30 to 50 % by weight. The aforesaid contents are expressed in "%" based on the total quantity of the components, excluding the additives described later, as 100. More specifically, they are expressed in terms of % by weight when the total quantity of the aforesaid ethylene- $\alpha$ ·olefin copolymer, the aforesaid SEPS, the aforesaid tackifying resin, and the plasticizer described later that is used as necessary is assumed to be 100. If the blending quantity of the aforesaid ethylene- $\alpha$ -olefin copolymer is less than 10 % by weight, the cohesive strength of the resulting hot melt adhesive decreases, which is not desirable. A quantity exceeding 80 % by weight causes the melt viscosity of the resulting hot melt adhesive to become extremely high, thus rendering itself undesirable. If the blending quantity of the aforesaid SEPS is less than 1 % by weight, miscibility deteriorates, whereas a quantity exceeding 50 % by weight could cause the lowtemperature adhesion property and texture to deteriorate, thus rendering itself undesirable. If the blending quantity of the aforesaid tackifying resin exceeds 60 % by weight, the texture becomes hard, and the low temperature adhesion property deteriorates. A quantity less than 19 % by weight is also not desirable because

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wettability to the adherend could decrease and deteriorate the initial adhesion strength.

[0038] In addition to the aforesaid ethylene- $\alpha$ -olefin copolymer, the aforesaid SEPS, and the aforesaid tackifying resin, the hot melt adhesive composition of the present invention may incorporate a plasticizer, as necessary. As the aforesaid plasticizer, the present invention can use any known plasticizers that are commonly used for hot melt adhesive compositions, with no specific limitation, and plasticizing oils can be cited as examples thereof. This type of plasticizer can be either liquid or solid. The blending quantity of the aforesaid plasticizer in the case of using it is preferably from 1 to 40 % by weight, better yet, from 5 to 30 % by weight, from the standpoint of the solidifying time of the adhesive and its tackiness after the solidification.

[0039] In addition to the aforesaid components, the hot melt adhesive composition of the present invention can further appropriately select and incorporate heretofore known additives, such as antioxidants, stabilizers, fillers, pigments, etc., as necessary. These additives can be used singly or in combination. Furthermore, besides the aforesaid additives, various kinds of thermoplastic macromolecular compounds can also be added.

[0040] The hot melt adhesive composition of the present invention can be prepared, for example, by placing the aforesaid raw materials

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in a melt mixing pot and by heating and mixing them and can be provided for use.

[0041] The hot melt adhesive composition of the present invention is useful as the adhesive used in the production of disposable diapers, catamenial pads, etc., (more specifically, for bonding various components together--for example, bonding a top sheet and back sheet.) In addition, it is suitably used in book binding, cardboard box production, and so forth.

# [0042] [Working Examples]

The following explains the present invention in more concrete terms by presenting working examples, but the present invention is not limited to or restricted by these. Incidentally, various properties mentioned in the working examples were determined by the following measuring methods.

# [0043] <Adhesion Property>

Polyethylene film (more specifically, a breathable sheet manufactured by Kao Co., 35  $g/m^2$ ), which is mainly used for paper diapers, and nonwoven cloth [more specifically, nonwoven cloth having a grammage of 20  $g/m^2$  that was comprised of polyethylene terephthalate (PET, core)/polyethylene (PE, sheath) core-in-sheath fibers] were obtained for the test. Said polyethylene film and nonwoven cloth were bonded by applying the hot melt adhesive composition obtained in a working example in a bead shape at a temperature of 120 to 150 °C with a coating quantity of  $0.05 \text{ g/m}^2$ , thereby obtaining a test piece. The

obtained test piece was left standing in a 20 °C temperature ambience for 24 hours and subsequently taken out, and the bonded components were pulled apart by hand. Here, "O" was assigned when material breakage occurred. "X" was assigned when the boundary separation occurred. " $\Delta$ " was assigned when the condition was between the two above. The bonding of the nonwoven cloth to nonwoven cloth was also evaluated in the same manner.

#### [0044] <Tackiness>

The hot melt adhesive composition obtained in a working example was applied onto 50  $\mu$ m-thick polyester film to a thickness of 50  $\mu$ m. This was cut to 25 mm x 125 mm, and this cut piece was formed into a loop so that the surface coated with the adhesive composition was on the exterior side, thus preparing a sample. This sample was bonded to a 25 mm-wide adherend (polyethylene plate) at a speed of 300 mm/minute and subsequently pulled upward at the same speed, under which state the force that separated the sample and the adherend was measured. /7 The measurement was taken twice, immediately after the application of the composition and 1 hour after the application. The average of five samples was taken as the measured value.

## [0045] <Hardness>

In a 20 °C temperature ambience, a hot melt adhesive composition that has been melted was poured into a ring having a diameter of 65 mm and a depth of 12 mm, and it was cooled and solidified, after which

its hardness was measured with a prescribed hardness tester (pursuant to JIS K6301).

#### <Seepage>

A hot melt adhesive composition was applied onto 50 mm [sic]—thick polyester film to a thickness of 50  $\mu$ m, thus obtaining a test sample. This sample was bonded with a commercially available porous polyethylene film having a grammage of 30 g/cm² and left standing at 60 °C for one week, after which the degree of seeping was visually evaluated. "O" was assigned when no seeping took place and the covering property of the film was achieved. "X" was assigned when the composition seeped out completely. " $\Delta$ " was assigned when the condition was between the two above.

### [0046] <Heat Stability>

## (1) Viscosity reduction rate

In a 180 °C temperature ambience, the hot melt adhesive composition obtained in a working example was left standing for 72 hours. The viscosity before and after that was measured, and the viscosity reduction rate was calculated and used as the index for heat stability. The formula used for this calculation is as follows: Viscosity reduction rate  $[(V_0 - V) / V_0] \times 100 (\%)$ 

V : Melt viscosity of the hot melt adhesive composition at 160 °C after it was left standing at 180 °C for 72 hours.

 $V_{\text{0}}$  : Melt viscosity of the hot melt adhesive composition at 160 °C prior to the heat history.

#### (2) Hue

The composition was left standing in a 180 °C temperature ambience for 72 hours, at which point the hue of the composition was visually evaluated. "O" was assigned when the composition was completely colorless or white. " $\Delta$ " was assigned when the composition was colored somewhat. "X" was assigned when the composition was colored clearly.

#### (3) Odor

The composition was left standing in a 180 °C temperature ambience for 72 hours, at which point the odor of the composition was evaluated by organoleptic tests (10 panelists). When there was no odor, "O" was assigned. When there was a slight smell, " $\Delta$ " was assigned. "X" was assigned when there was an obvious odor.

# [0047] [Working Examples]

According to each formula shown in Table 1, the components were mixed and further melted and kneaded at 150 °C, thereby formulating a hot melt adhesive composition. The aforesaid evaluations were carried out on the obtained hot melt adhesive compositions. The results are shown in Table 2.

[0048] [Table 1]

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エチレン		ン・ロ・ボレフィンは軽色は	904	-	2	c J	4	s	9	-	8	6	_
(MF	-	3 0 0 5	870g/s) (b)			1.チレン・1・ブラン水配合作(む)	1	エチレン・「・ヘキモン夫首合作(ロ)	Alek Cu	١	1977-1-1975/位部(3)	がは器の	-3
	_			0,7	35	2.0	=	1.5	0	3.0	30	35	_
		水母テルペン系生活 (d)	1977ロンアー1051(近原に対抗) (位にた) (位によ) (ので (の)	1	2.5	ı	0	1	1	40	1	2.0	·
5. 基整 (3. 基整		水松芳香栎条石油鄉 (a)	「アルコンP~100」(初1化学社会) 軟化点100℃(to	<b>4</b> 5	1	0 -	52	0.4	! !	1	1	9	
		水药缸防洗条石油盐烧 (f)	(イーストタックH-100W!(FARDSHMM)	1		3.0	]	ı	4.5	1	3.5	9 9	
		న జ న	「セブトン 2023」(ケラレ社的) Melt Index-1g/10…。 スチレン含有最-13% (4)		0_	i	2.5	1	1	0 -	1	1	~~~,~~~~
	- 1	7	セフトン 2013  (クラン社等)  Mclt   Index -   Tg/  fm;  スチレン会有量 -   3% (k)	vs	1	1.5		1.5	0 -	,	3.0	5	
हि	- ET	<b>迎利(1)</b>	「クリストール352」(エクソン化学社動(四)	30	30	2.5	9	9	2	00		18	
***	200	<b>酸化纺止原(n)</b>	ł	0.5	0.5	0.5	0.5		0.5	) e	G e	2 7	
**	Oze I	群外群吸仪剂 (p)	「チスピンP」(チバガイギーは20 (g)	0.3	0.3	0.3	9.3	0.3		3	2 6	. 0	
İ	;	€C	<b>i</b> (x)	100.8	8.8	8.0	18 8	88	8	ğ		2	
				-	-	-			<u> </u>	3 3	3	₹	

The blending quantities (numerical values) in the table indicate parts by weight.

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Key to Table 1: a) composition; b) ethylene- $\alpha$ -olefin copolymer (MFR = 300 g/cm<sup>2</sup>, density: 0.870 g/m); c) tackifying resin; d) hydrogenated terpene resin; e) hydrogenated aromatic petroleum resin; f) hydrogenated aliphatic petroleum resin; g) "Clearon P-105" (a product of Yasuhara Chemical Co.), softening point: 100 °C; h) "Alcon P-100" (a product of Arakawa Chemical Co.), softening point: 100 °C; i) "East Tac H-100W" (a product of Eastman Kodak Co.), softening point: 100 °C; j) "Septon 2023" (a product of Kuraray Co.) Melt Index: 1 g/10 min., styrene content: 13 %; k) "Septon 2043" (a product of Kuraray Co.) Melt Index: 17 g/10 min., styrene content: 13 %; 1) plasticizer; m) "Crystol 352" (a product of Exxon Chemical Co.); n) antioxidant; o) "Irganox 1010" (a product of Ciba Geigy Co.); p) ultraviolet absorber; q) "Chinubin P" (a product of Ciba Geigy Co.); r) total; s) working examples; t) ethylene - 1-butene copolymer; u) ethylene - 1-hexene copolymer; v) ethylene - 1-octene copolymer.

[0049] [Table 2]

			(T) 🗱		*		例		
	1	5	3	1	5	6	7	8	9
総職結度 (160℃, φs) <sub>(a)</sub>	1009	1万	7000	5000	3000	577	17.	2.5万	377
飲化点 (C)(b)	75	8 4	80	78	76	8 8	95	8 7	90
使 着 柱 PE+NW・	0	0	0	0	٥	0	Q	0	0
(\$ 0 (C) . NAX+NA	0	0	0	0	٥	0	0	0	0
(c) 強布直接 タックカ (20℃)	500	350	400	450	470	250	265	300	290
(点) (式) 時間後	500	340	400	440	460	250	260	800	285
しかはは全(0.0℃×1回回数)(g	, 0	0	0	0	0	0	0	С	0
M Ne変圧下率(a 1 1 6 0 °C)	10	6	7	8	9	2	3	5	4
至 18.0℃×72時収費の色程。 解	,0	٥	0	O	0	0	0	0	0
h) 180℃×12時間後の発気	, 0	0	0	0	0	0	0	0	0

\* In the table, PE stands for polyethylene film, and NW stands for nonwoven cloth.

Key: a) melt viscosity; b) softening point; c) adhesion property; d) tackiness; e) immediately after the application; f) 1 hour afterwards; g) seepage (60 °C x one week later); h) heat stability; i) viscosity reduction rate after 180 °C x 72 hours; j) hue after 180 °C x 72 hours; k) odor after 180 °C x 72 hours; 1) working examples; m) 10,000; n) 50,000; o) 40,000; p)25,000; q) 30,000.

[0050] [Effects of the Invention]

The hot melt adhesive composition of the present invention does not cause the seeping of low molecular substances, loses its tackiness after it is cooled and solidified, and, when used as the adhesive for assembling sanitary articles, such as disposable diapers, etc., does not impair the texture of said disposable diapers.